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HEAVY ELEMENTS

A DISSERTATION SUBMITTED TO THE
FACULTY OF THE DIVISION OF THE
PHYSICAL SCIENCES IN CANDIDACY FOR
THE DEGREE OF DOCTOR OF PHILOSOPHY

DEPARTMENT OF PHYSICS

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Packing Fraction Differences Among Heavy Elements

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A group of new packing-fraction-difference measurements are reported for the following elements: chromium, gadolinium, gold, lutecium, molybdenum, osmium, platinum, ruthenium, strontium, tantalum, tin, uranium, ytterbium and zirconium. By combining these differences with known packing fractions a number of new packing fractions are deduced. These are plotted on a packing-fraction curve. Atomic weights of osmium and lutecium are calculated. The atomic weight of osmium is still below the chemical value, while that of lutecium is shown to be the same as a recent chemical determination.

THE work of many investigators on mass spectra and nuclear disintegrations has established the shape and general aspects of the packing fraction curve. Many gaps exist in the curve for which no points have been measured and many of its details are masked by experimental errors. This work was undertaken in an attempt to check the shape and position of the curve and to establish accurate points in the region of heavy elements.

METHOD

If there are in the source of a mass spectrograph two isotopes of masses $m_1 = n_1 i_1 (1 + f_1)$ and $m_2 = n_2 i_2 (1 + f_2)$, m_1 and m_2 will form a doublet if $i_1 = i_2 = i$. In these expressions n_1 and n_2 are integers representing ionic charges, $n_1 i_1$ and $n_2 i_2$ are integers known as mass numbers and f_1 and f_2 are packing fractions defined by the above expressions. The mass difference between m_1 and m_2 can be determined from the separation of the members of this doublet if there are lines on the photograph to determine a mass scale. Let I and $I + k$, where k is a small integer, be the mass numbers of isotopes used for determining the mass scale. If $x - x'$ is the separation of these isotopes and $x_2 - x_1$ is the doublet separation, the required packing fraction difference is given by,¹

$$f_2 - f_1 = \frac{2(x_2 - x_1)}{i^{\frac{1}{2}}(x - x')} [(I + k)^{\frac{1}{2}} - I^{\frac{1}{2}}]. \quad (1)$$

It is preferable to bracket one isotope between two isotopes of a second element when no two sufficiently small charges, n_1 and n_2 , can be

found to satisfy the relation $n_1 i_1 / n_2 = n_2 i_2 / n_2$ (=mass/charge) or when isotopes with which the element can be doubled are lacking, rare, or for other reasons inconvenient. Moreover, the dissymmetry introduced in the field of view by the second member of a close doublet is known to influence the setting of a comparator cross hair on the first.² In bracketing, larger separations reduce this last effect to a minimum. By this method two isotopes of masses $n' i_1 (1 + f_1)$ and $n' i_2 (1 + f_2)$ appear as lines on either side of an isotope of a second element of mass $n i (1 + f)$ which appears on the plate at the position of mass $i(1 + f)$. If the positions of the lines are x_1 , x_2 and x , respectively, Dempster has shown that if we calculate,¹

$$a = \frac{(x - x_1)}{(x_2 - x_1)} (i_2^{\frac{1}{2}} - i_1^{\frac{1}{2}}) \quad (2)$$

$$\text{and} \quad \Delta m = (i^{\frac{1}{2}} + a)^2 - i, \quad (3)$$

$$\text{then} \quad \frac{\Delta m}{i} = f - \frac{(i_2^{\frac{1}{2}} - i_1^{\frac{1}{2}})}{(i_2^{\frac{1}{2}} - i_1^{\frac{1}{2}})} f_2 - \frac{(i_2^{\frac{1}{2}} - i_1^{\frac{1}{2}})}{(i_2^{\frac{1}{2}} - i_1^{\frac{1}{2}})} f_1. \quad (4)$$

For cases discussed in this paper Eq. (4) reduces to one of the following approximations.

$$\Delta m / i = f - \frac{1}{2}(f_1 + f_2), \quad (5)$$

$$\Delta m / i = f - \frac{1}{3}(2f_1 + f_2), \quad (6)$$

$$\Delta m / i = f - \frac{1}{4}(3f_1 + f_2). \quad (7)$$

Equation (5), (6) or (7) is selected depending upon whether the ratio of $x - x_1$ to $x_2 - x_1$ is approximately one-half, one-third or one-fourth,

¹ A. J. Dempster, Phys. Rev. **53**, 64 (1938).

² C. E. St. John and L. W. Ware, Astrophys. J. **44**, 17 (1916).

respectively. Approximations made in deriving the above formulae introduce no error larger than the experimental error.

PROCEDURE

Plates used in these determinations were taken on Dempster's double-focusing mass spectrograph³ and measured visually on a Gaertner comparator. The comparator, when checked for periodic error by measurements of a scale ruled on the ruling engine of Professor H. G. Gale, showed no appreciable error in the region of the screw used for these measurements. Microphotometer traces were taken of a number of exposures as a check on visual measurements. Since the screw of the microphotometer was not reliable, a trace of an accurate scale was made on the same film and doublet separations were compared with these known distances. Results obtained in this way checked the visual measurements but were not as self consistent and were not used in the results reported. For each set of elements studied, many exposures were taken and those to be measured were carefully selected on the basis of sharpness of focus, equality of intensity and symmetry of lines. This procedure accounts to a large extent for the accuracy obtained. Line intensities were equalized by varying the spark potential and by proper choice of an alloy.

Platinum-ruthenium-osmium

By using electrodes of an alloy of platinum, ruthenium and osmium in a high frequency arc source, excellent doublets were obtained at two masses. One at mass number 96 was singly charged ruthenium and doubly charged osmium; the other at mass number 99 was singly charged ruthenium and doubly charged platinum. Nineteen measurements were made on the first of these doublets and twenty-six on the second and packing fraction differences of $(+7.65 \pm 0.14)10^{-4}$ and $(+7.92 \pm 0.10)10^{-4}$, respectively, were obtained. With the same electrodes the platinum lines at 194, 195 and 196 were bracketed between ruthenium (96) and (99) and were measured on twelve, thirteen and twelve exposures, respectively. The differences obtained were $(+7.72$

$\pm 0.17)10^{-4}$, $(+7.71 \pm 0.20)10^{-4}$ and $(+7.69 \pm 0.20)10^{-4}$, respectively.

Platinum and osmium packing fractions

The packing fraction for the platinum line at 195 is known¹ to be $(+2.03 \pm 0.3)10^{-4}$. Hence, one can deduce the mean packing fraction of ruthenium (96) and (99) to be $(-5.68 \pm 0.36)10^{-4}$. If it is assumed that the weighted mean for these two elements as given in Eq. (6) is the same as that given in Eq. (5), the packing fractions for platinum (194) and (196) can be deduced. Results of this calculation are $(+2.04 \pm 0.4)10^{-4}$ and $(+2.01 \pm 0.4)10^{-4}$, respectively, which are nearly the same as Dempster's value for platinum (195). If the packing fraction of ruthenium (96) is assumed to be $(-5.68 \pm 0.36)10^{-4}$, the packing fraction of osmium (192) becomes $(+1.97 \pm 0.4)10^{-4}$. If in addition the platinum (194) value deduced above is assumed, the osmium (190) packing fraction becomes $(+2.00 \pm 0.4)10^{-4}$. In the table of masses submitted by the atomic committee of the International Union of Chemistry,⁴ the packing fraction of osmium is given as -1×10^{-4} . This is below the packing fraction curve and would place elements whose packing fractions are compared with osmium in this paper below the curve. Therefore the fraction for osmium should probably be raised. The atomic weight of osmium, with 2×10^{-4} for the packing fraction and Nier's⁵ value of the relative abundances for osmium, becomes 190.38. The agreement with the chemical value of 191.5 is better but leaves much to be desired.

Uranium-tin

With one electrode of uranium and one of tin, a doublet consisting of singly charged tin (119) and doubly charged uranium (238) was formed. Line intensities could be varied relative to each other by varying the spark voltage so that lines of equal intensity were obtained. The doublet was well separated and easily measured on twenty-six exposures. The mean value obtained was $(+10.12 \pm 0.09)10^{-4}$. The bracket of uranium (238) between tin (118) and (120) was measured on fourteen exposures as a check on the doublet

³ A. J. Dempster, *Phys. Rev.* **51**, 67 (1937).

⁴ O. Hahn, *Ber. d. D. Chem. Ges.* **71A**, 1 (1938).

⁵ A. O. Nier, *Phys. Rev.* **52**, 885 (1937).

measurement and was found to give a value of $(+10.12 \pm 0.13)10^{-4}$.

The known packing fraction for uranium,⁶ $(+5.56 \pm 0.1)10^{-4}$, gives for tin (119) the value $(-4.56 \pm 0.13)10^{-4}$ from the doublet, and for tin (118) and (120) an average value of $(-4.56 \pm 0.23)10^{-4}$ from bracket measurements. Dempster⁶ has given values for tin which are within the experimental errors given.

Gold-molybdenum

The gold-molybdenum bracket was obtained with one electrode of gold and one of molybdenum. The lines obtained were sharp and easy to measure. Eleven exposures of doubly charged gold (197) bracketed between molybdenum (97) and (100) were measured and checked by calculating the bracket of gold (197) between molybdenum (98) and (100) on twelve exposures. The mean differences obtained were $(+7.95 \pm 0.06)10^{-4}$ and $(+8.13 \pm 0.07)10^{-4}$, respectively.

The packing fraction for gold (197) is¹ $(+2.0 \pm 0.4)10^{-4}$ which gives as a mean packing fraction for molybdenum (97) and (100), $(-5.95 \pm 0.4)10^{-4}$ and for molybdenum (98) and (100) a mean value of $(-6.13 \pm 0.4)10^{-4}$. Dempster¹ has compared molybdenum with platinum at masses 97 and 98 and with osmium at masses 95 and 96. By using his results in conjunction with the values for platinum and osmium obtained in a previous section of this paper, the packing fractions of molybdenum (95), (96), (97) and (98) are found to be $(-5.76 \pm 0.5)10^{-4}$, $(-5.61 \pm 0.5)10^{-4}$, $(-5.66 \pm 0.5)10^{-4}$ and $(-5.67 \pm 0.5)10^{-4}$, respectively.

Tantalum-zirconium

Doubly charged tantalum (181) was bracketed between singly charged zirconium (90) and (91) when metallic electrodes of these elements were used. Six exposures were measured to determine a mean packing fraction difference of $(+7.71 \pm 0.09)10^{-4}$. The bracket of tantalum (181) between zirconium (90) and (92) was also measured on six exposures. The difference obtained from this bracket was $(+7.77 \pm 0.08)10^{-4}$.

Aston⁷ gives an estimate of the packing fraction of tantalum of $(-4 \pm 3)10^{-4}$. This value is

⁶ A. J. Dempster, *Phys. Rev.* **53**, 872 (1938).

⁷ F. W. Aston, *Mass Spectra and Isotopes* (Edward Arnold & Co., 1933), pp. 141, 235.

about 5 units below the curve and would place zirconium about the same distance below the curve. This is improbable and no attempt to deduce a packing fraction for either of these elements will be made until a more accurate value for one of them has been obtained.

Gadolinium-chromium

Many exposures with different types of electrodes were taken of these two elements in getting equal intensities. The large number of electrodes used permitted four different measurements to be made with optimum intensities for each. For one set of plates an electrode was made by packing a nickel tube with gadolinium oxide and filings of neodymium. The second electrode was of Nichrome wire for some plates and stainless steel for others. Other plates were made with both electrodes of nickel tubes filled with 25 mg of gadolinium oxide, 25 mg of lanthanum and from 0.5 to 5.0 mg of chromium filings. Eighteen measurements were made on the triply charged gadolinium (156), singly charged chromium (52) doublet. This measurement was checked by the bracket of chromium (52) between gadolinium (155) and (157) on sixteen exposures. The differences obtained were $(+6.37 \pm 0.11)10^{-4}$ and $(-6.39 \pm 0.09)10^{-4}$, respectively. Chromium (53) was bracketed between gadolinium (158) and (160) on ten exposures and on six exposures gadolinium (160) was bracketed between chromium (53) and (54). The results measured on these brackets were $(-6.38 \pm 0.13)10^{-4}$ and $(+6.33 \pm 0.26)10^{-4}$, respectively.

Recently Aston⁸ has reported a value for the packing fraction of chromium (52) of -8.18×10^{-4} with an error in mass units of 0.0008 which corresponds to an error in his packing fraction of 0.15×10^{-4} . If this value is assumed, the packing fraction of gadolinium (156) is $(-1.81 \pm 0.19)10^{-4}$ and the average value for gadolinium (155) and (157) is $(-1.79 \pm 0.18)10^{-4}$. The assumed packing fraction of chromium and those values deduced from it are below the packing fraction curve.

Ytterbium-strontium

For ytterbium and strontium determinations, electrodes were nickel tubes filled with 25 mg

⁸ F. W. Aston, *Nature* **141**, 1096 (1938).

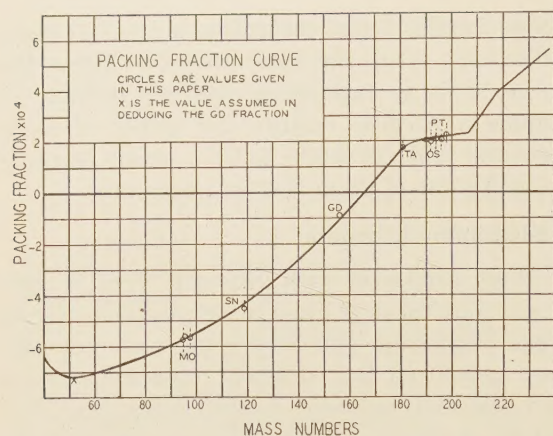


FIG. 1.

of ytterbium oxide, 25 mg of lanthanum and varying amounts of strontium. Plates were taken using 10, 15, 30 and 60 mg of strontium filings. Twenty-four measurements were made of the doublet formed by doubly charged ytterbium (172) and singly charged strontium (86) to give a mean packing fraction difference of $(+6.20 \pm 0.15)10^{-4}$. Ten measurements of the doublet between ytterbium (174) and stron-

tium (87) were made with a mean result of $(+6.18 \pm 0.14)10^{-4}$. In addition strontium (86) was bracketed between ytterbium (171) and (173) on thirteen exposures. The measured difference was $(-6.48 \pm 0.19)10^{-4}$. A strong line was found at the position of mass 175. The only isotope of this mass number is lutecium which was present as an impurity in the ytterbium. Seven measurements were made on the strontium (87) line bracketed between ytterbium (173) and lutecium (175) and the difference measured was $(-6.37 \pm 0.16)10^{-4}$. A comparison of this result with other results discussed in this section indicates that there can be no large difference in the packing fractions of lutecium and ytterbium. This was checked by making ten measurements of the lutecium line compared with ytterbium (173) and (174). The results of this and a different calculation on the same measurements were $(0.00 \pm 0.34)10^{-4}$ and $(+0.05 \pm 0.20)10^{-4}$, respectively.

That the packing fraction of lutecium is about the same as that of ytterbium is of interest in view of the recent atomic weight determination

TABLE I. Summary of doublets and brackets measured. I is the mass number at which the comparison is made; n is the number of exposures measured; $f = (m - I)/I$ is the packing fraction if m is the mass and I is the mass number of the ion. In the description of the function measured in the last column, the f 's are in order of the ions in column 3.

COMPARISON NUMBER	I	IONS COMPARED (PART 1)	n	$(f_1 - f_2) 10^4$
1	52	Gd(156) - Cr(52)	18	$+6.37 \pm 0.11$
2	86	Yb(172) - Sr(86)	24	$+6.20 \pm 0.15$
3	87	Yb(174) - Sr(87)	10	$+6.18 \pm 0.14$
4	96	Os(192) - Ru(96)	19	$+7.65 \pm 0.14$
5	99	Pt(198) - Ru(99)	26	$+7.92 \pm 0.10$
6	119	U(238) - Sn(119)	26	$+10.12 \pm 0.09$
(PART 2)				$[f_1 - \frac{1}{2}(f_2 + f_3)] 10^4$
7	52	Cr(52) - Gd(155), Gd(157)	16	-6.39 ± 0.09
8	53	Cr(53) - Gd(158), Gd(160)	10	-6.38 ± 0.13
9	86	Sr(86) - Yb(171), Yb(173)	13	-6.48 ± 0.19
10	87	Sr(87) - Yb(173), Lu(175)	7	-6.37 ± 0.16
11	90.5	Ta(181) - Zr(90), Zr(91)	6	$+7.71 \pm 0.09$
12	96	Ru(96) - Os(190), Pt(194)	5	-7.70 ± 0.11
13	97.5	Pt(195) - Ru(96), Ru(99)	13	$+7.71 \pm 0.20$
14	98.5	Au(197) - Mo(97), Mo(100)	11	$+7.95 \pm 0.06$
15	119	U(238) - Sn(118), Sn(120)	14	$+10.12 \pm 0.13$
16	174	Yb(174) - Yb(173), Lu(175)	10	$+0.05 \pm 0.20$
(PART 3)				$[f_1 - \frac{1}{3}(2f_2 + f_3)] 10^4$
17	53.3	Gd(160) - Cr(53), Cr(54)	6	$+6.33 \pm 0.26$
18	97	Pt(194) - Ru(96), Ru(99)	12	$+7.72 \pm 0.17$
19	98	Pt(196) - Ru(99), Ru(96)	12	$+7.69 \pm 0.20$
20	175	Lu(175) - Yb(174), Yb(173)	10	$+0.00 \pm 0.34$
(PART 4)				$[f_1 - \frac{1}{4}(3f_2 + f_3)] 10^4$
21	90.5	Ta(181) - Zr(90), Zr(92)	6	$+7.77 \pm 0.08$
22	98.5	Au(197) - Mo(98), Mo(100)	12	$+8.13 \pm 0.07$

TABLE II. *New packing fraction determinations. By using packing fraction differences given in Table I and assuming a packing fraction for one element, the packing fraction for a second element can be determined. The comparison numbers of the first column correspond to those in Table I.*

COMPARISON NUMBER	$f \times 10^4$ ASSUMED		$f \times 10^4$ DEDUCED	
13	Pt(195)	$+2.03 \pm 0.3$	Ru(96), (99)	-5.68 ± 0.36
18	Ru(96), (99)	-5.68 ± 0.36	Pt(194)	$+2.04 \pm 0.4$
19	Ru(96), (99)	-5.68 ± 0.36	Pt(196)	$+2.01 \pm 0.4$
5	Ru(99)	-5.68 ± 0.36	Pt(198)	$+2.24 \pm 0.4$
12	Ru(96)	-5.68 ± 0.36		
	Pt(194)	$+2.04 \pm 0.4$	Os(190)	$+2.00 \pm 0.4$
4	Ru(96)	-5.68 ± 0.36	Os(192)	$+1.97 \pm 0.4$
6	U(238)	$+5.56 \pm 0.1$	Sn(119)	-4.56 ± 0.13
15	U(238)	$+5.56 \pm 0.1$	Sn(118), (120)	-4.56 ± 0.23
14	Au(197)	$+2.00 \pm 0.4$	Mo(97), (100)	-5.95 ± 0.4
22	Au(197)	$+2.00 \pm 0.4$	Mo(98), (100)	-6.13 ± 0.4
Mo-Os ¹	Os(190)	$+2.00 \pm 0.4$	Mo(95)	-5.76 ± 0.5
Mo-Os ¹	Os(192)	$+1.97 \pm 0.4$	Mo(96)	-5.61 ± 0.5
Mo-Pt ¹	Pt(194)	$+2.04 \pm 0.4$	Mo(97)	-5.66 ± 0.5
Mo-Pt ¹	Pt(196)	$+2.01 \pm 0.4$	Mo(98)	-5.67 ± 0.5
1	Cr(52)	-8.18 ± 0.15	Gd(156)	-1.81 ± 0.19
7	Cr(52)	-8.18 ± 0.15	Gd(155), (157)	-1.79 ± 0.18

of Hönigschmid⁹ on lutecium. He obtained an atomic weight of 174.98 and by assuming a packing fraction of -4×10^{-4} he deduced an atomic weight from physical measurements of 174.91. When this was corrected for an isotope reported by Gallnow¹⁰ from spectroscopic evidence to be present at 173 or 177, he obtained an atomic weight of 174.96 if the higher isotope was present to about 2.5 percent. From the present curve it is improbable that the packing fraction of lutecium is lower than 0.5×10^{-4} . This corresponds to an atomic weight of 174.96 and if there is an isotope present at 177 its relative abundance must be less than one percent.¹¹

SUMMARY

Results discussed above are summarized in Tables I and II, and in Fig. 1. Table I is divided into four parts. Part 1 gives the results obtained when the function of the packing fraction given by Eq. (1) is calculated. Similarly, Parts 2, 3 and 4 are calculations from Eqs. (5), (6) and (7). Plus and minus values appended are probable errors.

The author wishes to express his appreciation to Professor A. J. Dempster for permission to use his mass spectrograph, for suggesting the problem and for his many helpful suggestions in completing the above investigation.

relative abundance of 2.58 percent. This yields an atomic weight of 174.99 in good agreement with Hönigschmid's value.

⁹ O. Hönigschmid, *Naturwiss.* **25**, 748 (1937).

¹⁰ H. Gallnow, *Zeits. f. Physik* **103**, 443 (1936).

¹¹ J. Mattauch and H. Lichtblau (*Zeits. f. Physik* **111**, 514 (1939)) report an isotope of lutecium at 176 with a



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